

# **Tensile Properties and Scanning Electron Microscopy Examination of the Fracture Surface of Oil Palm Wood Flour/Natural Rubber Composites**

Hanafi Ismail and Hasliza Isa Nurdin

School of Industrial Technology, Universiti Sains Malaysia, 11800 Minden, Penang, Malaysia

Received: 19 October 1997; accepted 28 February 1998

## **ABSTRACT**

Tensile properties and scanning electron microscopy examination of the fracture surface of oil palm wood flour/natural rubber composites have been studied. Tensile modulus of the composites increases with oil palm wood flour concentration, whereas, tensile strength and elongation at break show the opposite trend. The adhesion between oil palm wood flour and rubber matrix can be enhanced by the use of various bonding agents. The improvement in interfacial interaction between oil palm wood flour and rubber is substantiated by means of scanning electron microscopy.

**Key Words:** oil palm, tensile properties, natural rubber, scanning electron microscopy, bonding agents

## **INTRODUCTION**

The reinforcement of a rubber with fibres combines the elastic behaviour of the rubber with the strength and stiffness of the reinforcing fibre. Although the use of short fibres as a filler in rubber compound was started long ago but only lately short fibre-reinforced has gained importance because of the advantage of better processing, high anisotropy in mechanical properties, excellent stiffness characteristics, good adhesion to rubber matrix and lower cost. Previous works on this topic have been reported by numerous researchers [1-7].

In our previous works [8, 9], we have reported the curing characteristics and mechanical properties of oil palm fibre reinforced natural rubber composites. Mechanical properties of the composites were

enhanced with the modification on fibre surface and the use of various bonding agents. The presence of bonding agents in composites prolonged the curing time. This paper highlights the tensile properties and scanning electron microscopy studies of the fracture of oil palm wood flour (OPWF)/natural rubber composites. Emphasis has been given to the effects of OPWF loading and bonding agents on the tensile properties.

## **EXPERIMENTAL**

Natural rubber (SMR L) was obtained from the Rubber Research Institute of Malaysia (RRIM). Oil palm empty fruit bunch (EFB) in fibrous form used in this study was obtained from Sabutek (M) Ltd, Perak,

**Table 1.** Formulation of the mixes (composition in 100 parts by weight of rubber).

| Composite                     | A   | B   | C   | D   | E   | F   | G   |
|-------------------------------|-----|-----|-----|-----|-----|-----|-----|
| SMRL                          | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| OPWF                          | -   | 15  | 30  | 50  | 15  | 15  | 15  |
| Phenol formaldehyde (PF)      | -   | -   | -   | -   | 10  | -   | -   |
| Resorcinol formaldehyde (RF)  | -   | -   | -   | -   | -   | 5   | 5   |
| Silica (Sil)                  | -   | -   | -   | -   | -   | -   | -   |
| Hexamethylenetetramine (Hexa) | -   | -   | -   | -   | -   | 2   | 2   |
|                               | -   | -   | -   | -   | -   | -   | 5   |

All the mixes contain stearic acid 2; zinc oxide 5; sulphur 2.5; CBS 0.6; Flectol H 1.

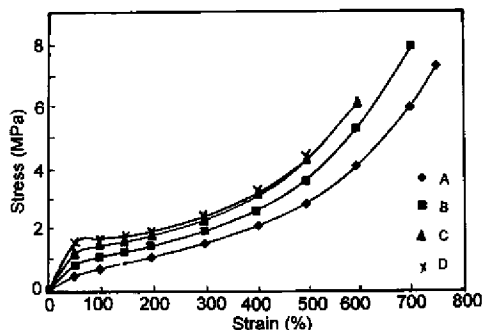
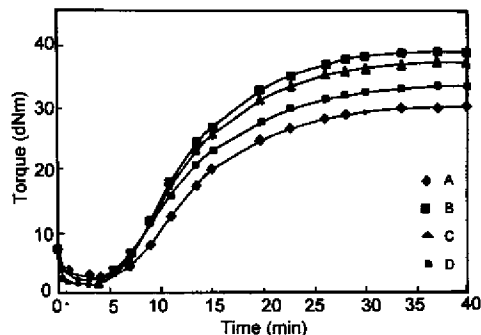
Malaysia and were ground into sizes of 180–270  $\mu\text{m}$ . The formulations of the mixes are given in Table 1. Sulphur, zinc oxide, stearic acid, *N*-cyclohexylbenzothiazyl sulphenamide (CBS) and silica were all purchased from Bayer (M) Ltd. Poly-1,2-dihydro-2,2,4-trimethyl quinoline (Flectol H) was supplied by Monsanto Company (M). Bonding agents used were: phenol formaldehyde from Borden Chemical (M) Ltd, hexamethylenetetramine from Fluka Chemical (M) Ltd, and resorcinol formaldehyde from Lianco (M) Ltd.

The composites were prepared on a conventional laboratory two roll mill size according to ASTM designation D3184–80. Nip gap, mill roll speed ratio, time of mixing, and the sequence of addition of the ingredients were kept the same for all the composites. Sheets of 2 mm thickness were moulded in an electrically heated hydraulic press at

obtained from a Monsanto R–100 rheometer. Tensile specimens were punched out along the grain direction and were tested on Instron universal testing machine, Model 1114, according to ASTM D412 at 500 mm/min cross-head speed. The fracture surfaces were sputter coated with gold within 72 h of testing and were observed using a Leica Cambridge S–360 Model scanning electron microscope.

For swelling measurement the cured test pieces of dimensions 30 $\times$ 5 $\times$ 2 mm were swollen in toluene until equilibrium swelling was achieved, which normally took 72 h at 25  $^{\circ}\text{C}$ . In this study, *Q* (the weight of toluene uptake per gram of rubber hydrocarbon) was determined according to the expression derived by Park and Brown [10] as shown below.

$$Q = \frac{\text{swollen} - \text{deswollen wt}}{\text{dry wt} \times 100 / \text{formula wt}}$$

**Figure 1.** Stress-strain behaviour of the composites A–D.**Figure 2.** Rheographs of the composites A–D at 150  $^{\circ}\text{C}$ .

**Table 2.** Tensile properties of the composites.

| Composite                              | A    | B    | C   | D   | E    | F    | G    |
|--|------|------|-----|-----|------|------|------|
| Modulus at 100% elongation, M100 (MPa) | 0.7  | 1.1  | 1.4 | 1.7 | 1.3  | 1.1  | 1.4  |
| Modulus at 300% elongation, M300 (MPa) | 1.6  | 1.9  | 2.3 | 2.4 | 2.3  | 2.1  | 2.7  |
| Tensile strength (MPa)                 | 19.0 | 11.6 | 7.4 | 5.4 | 13.7 | 12.8 | 16.5 |
| Elongation at break (%)                | 813  | 784  | 673 | 568 | 776  | 732  | 674  |

The rubber-filler interaction was calculated as suggested by Lorenz and Park [11].

$$\frac{Q_r}{Q_s} = ae^{-1} + b$$

The subscripts f and g refer to filled and gum vulcanizates, respectively. Z is the ratio by weight of filler to rubber hydrocarbon in the vulcanizate, while a and b are constants.

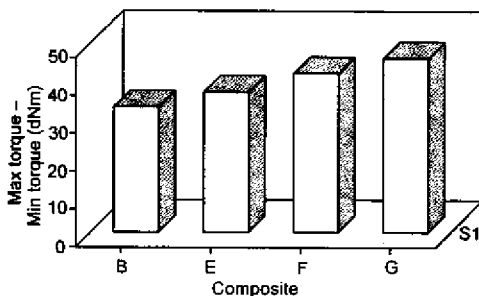
## RESULTS AND DISCUSSION

Figure 1 represents the stress-strain curves of the composites A–D. In this figure the stress-strain curves exhibit on the well known sigmoidal non-linear characteristic of rubber like materials. Initially a rapid stress growth takes place especially at less than 100% elongation. This may reflect the existence of strong OPWF/rubber adhesion that restricts the deformability of the composites. However, at higher elongation, the separation of OPWF from the rubber matrix will eventually lead to the failure of the composites. Addition of fibre increases the modulus of the composites. This observation can be seen also from the rheographs of the composites A–D at 150 °C in Figure 2. The modulus of the composites increases as shown by the increment of the maximum torque with OPWF loading.

The tensile properties of all composites studied are shown in Table 2. It can be seen that the tensile strength decreases with increasing OPWF loading. As the OPWF loading is increased, the reinforcing effect brought about is insufficient to compensate for the dilution effect of the OPWF in the rubber matrix and hence a decrease in tensile strength is observed.

The elongation at break shows a reduction with increasing OPWF loading. According to Hewitt [12] and Lorenz and Park [11], the incorporation of fibre in rubber matrix reduces resilience and toughness of the composites and leads to lower resistance to break. As mentioned before, the incorporation of OPWF increases the modulus of the composites.

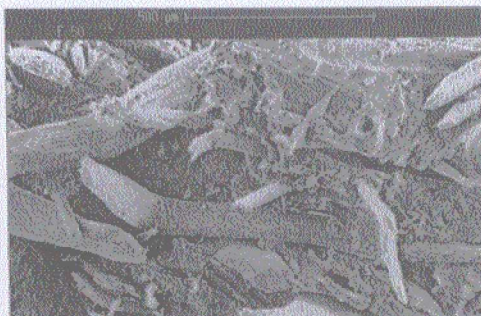
Table 2, also, indicates that the use of various bonding systems improve the tensile properties of the composites. Figure 3 shows the maximum torque–minimum torque comparison between composites B (15 phr of OPWF but without bonding agent) and composites with different types of bonding agent at 15 phr of OPWF. Our previous works [8, 9] have shown that the bonding between the fibre and rubber matrix is improved with the addition of bonding agents in the rubber composites. The strong adhesion between fibre and matrix resulted in higher shear strength at the interface and stronger force must be used to overcome the shear strength at the interface which resulted in a higher tensile strength and tensile



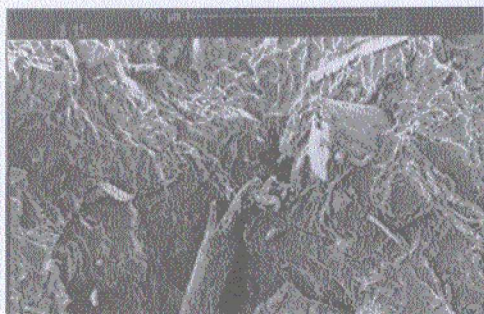
**Figure 3.** Relationship between maximum torque–minimum torque and filler loading of the OPWF filled natural rubber composites.



a



d



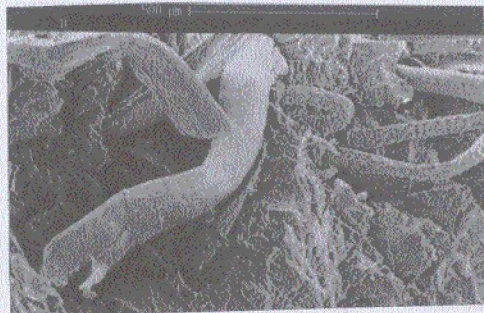
b



e



c



f

**Figure 4.** SEM micrographs of the OPWF filled natural rubber composites after tensile fracture at different filler loading (a) 0 phr (control composite); (b) 15 phr; (c) 30 phr; (d) 50 phr; (e) 15 phr with bonding agent (composite E); (f) 15 phr with bonding agent (composite G) at magnification  $\times 70$ .

modulus [13].

### Scanning Electron Microscopy Studies

According to Kumar and Thomas [14], SEM is an important tool for observing the surface morphology of fibres, the cause of crack initiation and the failure process in composite materials. Figures 4a–4f show the SEM photomicrograph of the fracture surfaces of tensile test specimen of the composites studied. The fracture surface of natural rubber composite without OPWF (composite A) in Figure 4a exhibits brittle fracture. However, the presence of OPWF changes the failure pattern which indicates the presence of debonded OPWF as a result of poor OPWF/rubber interaction (Figures 4b–4d). As the OPWF loading increases, a greater number of debonded OPWF and a large number of holes can be seen as a result of the pull-out of OPWF from the rubber matrix. The decreased wettability and a greater number of OPWF in Figures 4b–4d are quite insufficient for uniform transmission of stress in the matrix resulting in localized stresses at the fibre–matrix interface leading to the reduction of tensile properties viz tensile strength and elongation at break.

Figures 4e–4f show the fracture surfaces of the composites with different types of bonding agents. Compared to Figure 4b (composite B) which contains no bonding agent but similar loading of OPWF (15 phr), it can be seen that composites in Figures 4e–4f (with bonding agents) show better filler–rubber adhesion. The stronger OPWF/rubber adhesion in Figures

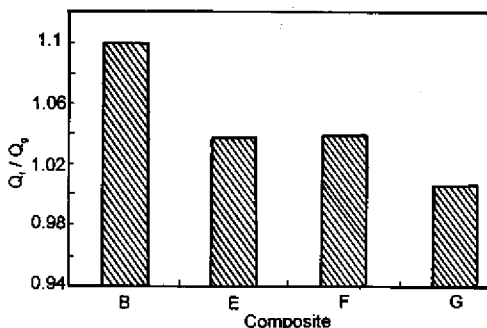


Figure 5. The effect of different bonding agents on  $Q_f/Q_m$  of OPWF filled natural rubber composites.

4e–4f causes breakage of the fibres without pulling them out of the rubber matrix. This is due to the increase in adhesion between the rubber and OPWF through bonding agents. Both figures also show that there are very low pull-out of fibres on the fracture surfaces. This observation is supported by the swelling measurement data shown in Figure 5. Compared to composite B, the addition of various bonding agents in composites E, F and G gives lower  $Q_f/Q_m$  value. Since the lower the  $Q_f/Q_m$  values, the higher will be the extent of interaction between the filler and the rubber matrix [11], it can be concluded that the use of various bonding agents has increased the OPWF/rubber adhesion in the composites. This explains why the tensile properties of OPWF filled natural rubber composites with bonding agents are higher than the similar composite but without bonding agent.

### CONCLUSION

OPWF has a potential to be used as a cheap, low-active filler in the rubber products which do not require high strength and which are not used for dynamic applications. The adhesion between OPWF and rubber matrix is poor and can be enhanced by the use of suitable bonding agents.

### ACKNOWLEDGEMENT

The supply of oil palm empty fruit bunch (EFB) by Sabutek (M) Ltd is gratefully acknowledged. The authors wish to express their thanks to Universiti Sains Malaysia (USM) for the financial support.

### REFERENCES

1. Derringer G.C., *Rubb. World*, **165**, 45, 1975.
2. O'Connor J.E., *Rubb. Chem. Technol.*, **50**, 945, 1977.
3. Coran A.Y., *Rubb. Chem. Technol.*, **47**, 396, 1974.
4. Coran A.Y., Hamed P., and Goettler L.A., *Rubb. Chem. Technol.*, **49**, 1167, 1976.

5. Remy M.M. and De B.K., *Rubb. Chem. Technol.*, **56**, 287, 1982.
6. Remy M. and Simeoni G., *J. Appl. Polym. Sci.*, **43**, 1805, 1994.
7. Prasanna K., Subhakarani A.M.L., and Sabu T., *J. Appl. Polym. Sci.*, **50**, 297, 1993.
8. Hanafi Ismail, Ramli N., and Rozman H.D., *Polymer*, **38**, 4059, 1997.
9. Hanafi Ismail, Ramli N., and Rozman H.D., *Eur. Polym. J.*, **33**, 1231, 1997.
10. Park C.P. and Brown R.J., *Rubb. Chem. Technol.*, **49**, 233, 1976.
11. Lorenz O. and Paris C.R., *J. Polym. Sci.*, **50**, 295, 1961.
12. Hewitt N.L., *Rubb. Age*, **1**, 59, 1972.
13. Miwa M., Ohsawa T., and Tshara K., *J. Appl. Polym. Sci.*, **25**, 795, 1980.
14. Prasanna Kumar R. and Sabu T., *Polym. Int.*, **38**, 173, 1995.